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Significant contribution from impurity-band transport to the room temperature conductivity of silicon-doped AlGa_N

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Abstract: Silicon-doped *n*-type (0001) AlGa_N materials with 60% and 85% AlN content were studied close to the doping condition that gives the lowest resistivity (Si/III ratios in the ranges $2.8\text{--}34 \times 10^{-5}$ and $1.3\text{--}6.6 \times 10^{-5}$, respectively). Temperature-dependent conductivity and Hall-effect measurements showed that, apart from the diffusion-like transport in the conduction band, a significant amount of the conductivity was due to phonon-assisted hopping among localized states in the impurity band, which became almost completely degenerate in the most doped sample of the Al_{0.6}Ga_{0.4}N series. In the doping range explored, impurity-band transport was not only dominant at low temperature, but also significant at room-temperature, with contributions to the total conductivity up to 46% for the most conductive sample. We show that, as a consequence of this fact, the measurements of Hall carrier concentration and Hall mobility using the usual single-channel approach are not reliable, even at high temperatures. We propose a simple method to separate the contributions of the two channels. Our model, although only approximate, can be used to gain insight into the doping mechanism: particularly it shows that the room-temperature free-electron concentration in the conduction band of the Al_{0.6}Ga_{0.4}N material reaches its maximum at about $1.6 \times 10^{18} \text{ cm}^{-3}$, well below the value that would have been obtained with the standard single-channel analysis of the data. This maximum is already achieved at dopant concentrations lower than the one that gives the best conductivity. However, further increase of the doping levels are required to enhance the impurity-band channel, with concentrations of the carriers participating in this type of transport that increase from $2.1 \times 10^{18} \text{ cm}^{-3}$ up to $4.3 \times 10^{18} \text{ cm}^{-3}$. For the Al_{0.85}Ga_{0.15}N, even though it was not possible to estimate the actual carrier concentrations, our measurements suggest that a significant impurity-band channel is present also in this material.

Keywords: III-nitride materials, AlGa_N, doping, hopping conduction.

1. Introduction

In recent years, the *n*-type doping of AlGa_N alloys has been extensively studied by many groups especially for their use as cladding layers in ultraviolet light emitting diodes. However, doping of those materials has proven to be very difficult, especially for the high aluminium concentration range [1]. Early work [2, 3] in this field suggested that quite a high dopant incorporation is necessary in order to achieve a reasonable level of conductivity, but this requirement is in contrast with the necessity to avoid the onset of severe self-compensation. More recently, Mehnke *et al.* [4] have investigated this trade-off condition for aluminium-rich

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alloys, and they have shown that there is an optimum doping condition, expressed in terms of partial pressure ratio of the silicon containing gas over the metal precursors during growth (Si/III ratio), that gives the best material. Similar behaviour was also reported for semipolar (11 $\bar{2}2$) *n*-AlGa \bar{N} with AlN concentration around 50% [5]. However, the physical reasons for that are still not fully understood.

2. Experimental

In this letter, in order to shed some light into this mechanism, we performed Hall-effect and conductivity measurements using the van der Pauw configuration. The Al_{0.6}Ga_{0.4}N material, which was co-loaded with the semipolar samples already reported elsewhere [5], was grown by MOVPE with a V/III ratio of 600 and a growth pressure of 50 mbar. The Al_{0.85}Ga_{0.15}N material was grown at similar conditions but with a V/III ratio of 460. In both cases, the growth was performed on AlN templates previously obtained using our standard procedure [6] on top of sapphire substrates; the nominal thickness of the doped layers was 1050 nm for the Al_{0.6}Ga_{0.4}N material and 800 nm for the Al_{0.85}Ga_{0.15}N material. The wafers were then diced into 10 × 10 mm² squares, and metal contacts were evaporated on the corners to form the samples. The metal stack was a sequence of Ti/Al/Ti/Au with thicknesses of 20/150/50/100 nm, respectively. The contacts were annealed in N₂ ambient for 1 minute at 800°C. Conductivity and Hall coefficient were acquired using a Lake Shore HMS 8404 equipment, with a closed cycle refrigerator unit for temperature regulation from 10 K to 400 K. The measurements were performed in dark condition with a magnetic field of 0.9 T and a current that varied from 100 μ A to 10 mA depending on the resistivity of the samples. The two most highly doped samples of the 60% AlN-content series appeared severely compensated and showed a very different behaviour; they are not included in this analysis and will be discussed elsewhere [7].

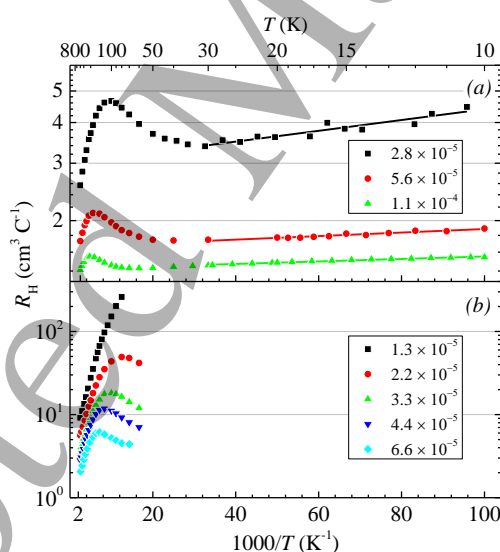


Figure 1. Temperature dependence of Hall coefficient for (a) *n*-Al_{0.6}Ga_{0.4}N and (b) *n*-Al_{0.85}Ga_{0.15}N grown with different Si/III ratios.

3. Results and discussion

As it is customary, we express the results of the Hall-effect measurements in terms of the Hall coefficient (R_H), defined as

$$R_H = \frac{E_t}{J B_{\perp}} \quad (1)$$

where J is the current density into the sample, B_{\perp} the magnetic field applied perpendicularly to the current, and E_t the resulting transverse electric field caused by the Hall effect. The logarithm of R_H vs the inverse of temperature is hence reported in figure 1 for (a) $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ and (b) $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$, respectively.

In case of the $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ samples, three regimes are clearly apparent: a high temperature one, in which R_H increases exponentially, as expected in case of conductivity due to electrons thermally activated into the conduction band, an intermediate temperature one in which R_H reaches its maximum, and finally a low temperature one in which R_H stays almost constant showing a very small, although well defined, thermal activation behaviour, with energies that decrease as the doping level increases. For the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ set of samples, it was not possible to have reliable measurements at temperatures lower than 60–70 K as the contacts became non-ohmic. However, in the range of temperatures explored, the presence of a maximum in R_H was also clearly visible in all the samples apart from the least doped one. Moreover, the samples showed the same trend as in $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$, in which the R_H maximum shifts towards higher temperatures as the doping level is increased.

A variation of the Hall coefficient with temperature similar to that shown in figure 1 has already been reported for germanium [8], and gallium nitride [9–11], and was explained as due to the simultaneous presence of two conduction channels: the usual band transport and some form of impurity conduction with a much lower mobility. The Hall coefficient can then be fitted using the usual formula for mixed conductivity

$$R_H = \frac{R_{con}\sigma_{con}^2 + R_{imp}\sigma_{imp}^2}{(\sigma_{con} + \sigma_{imp})^2}, \quad (2)$$

in which R_{con} and R_{imp} are, in this case, the Hall coefficients of the band-like channel and the low-mobility channel, respectively; and σ_{con} and σ_{imp} are their respective conductivities. It is important to notice that the standard conduction band and the so-called impurity band that forms in highly-doped semiconductor materials have radically different structures. While the former contains delocalized states extending to the whole crystal, the term impurity-band refers to a statistical distribution of states, each of them localized to a different donor impurity. This difference reflects on the type of transport that the two bands can support: diffusive for the carrier thermally activated to the conduction band, and phonon-assisted tunnelling among localized states, or so-called hopping conduction, in the impurity band. Only when the superposition among the donor waveforms is very large, which happens at even higher doping levels, does conduction also start to become diffusive in the impurity band. Such a metal transition in the impurity band has been convincingly reported e.g. in [12], where it was observed in n -doped germanium or, more recently, in [13], where this regime was closely approached in p -doped silicon carbide.

Although the mobility is always higher in the conduction band than in the impurity band, the former can be the dominant channel only at high temperatures, when enough carriers are thermally activated to the delocalized states. On the other hand, conductivity due to hopping transport is considerably less sensitive to temperature and, although it also decreases with temperature, it eventually becomes the dominant transport mechanism when the free-carrier concentration in the conduction band is negligible. For this reason, at high and low temperatures

when σ_{con} or σ_{imp} dominates the total conductivity, Eq. (2) reduces to R_{con} and R_{imp} , respectively. At intermediate temperatures, on the other hand, the switchover from one channel to the other as the dominant transport mechanism can be delayed if the ratio of the two mobilities is very high and a maximum might indeed appear as in the data of figure 1.

Even though this behaviour of R_H is not new in other highly-doped semiconductor materials, to the best of our knowledge this is the first time that it has been reported for AlGa_{0.4}N. Moreover, because of the particularities of this alloy, none of the methods proposed in the previously cited works regarding Ge and GaN [8-10] can be used with our material to separate the contributions of the two channels.

In the work of Hung and Gliessman [8] the authors suggest that R_{con} and R_{imp} can be simply isolated at high and low temperatures. While this seems to be reasonable in order to isolate R_{imp} at low temperatures (i.e. in the 10–30 K range), in which we can assume no significant band conduction, the same is not necessarily true for R_{con} as we cannot rule out the possibility of having considerable impurity conduction even at temperatures as high as those we have explored here. Specifically in order to respond to this objection, Molnar *et al.* [9] and later Look *et al.* [10] have developed more elaborate models that can effectively be used to separate the two contributions in GaN samples, provided some initial assumptions are met. In particular, the Molnar model assumes that the total number of carriers in the two bands is constant, which implies that the impurity band must be completely degenerate with no possible sign of thermally activated behaviour. The Look model assumes that the uncorrected mobility calculated using a single-channel model should asymptotically approach the almost constant impurity-band mobility at low temperature. However, neither of those two assumptions holds for our material as thermally activated behaviour is clearly present in the impurity conduction, and the uncorrected mobility keeps decreasing as the temperature is lowered without stabilizing to any asymptotic value, as will be shown later (see figure 4).

To overcome these problems and effectively estimate the contributions of the two conductivity channels in our material, we propose here a new method. The fact that at low temperature the logarithm of R_H is a linear function of the inverse of the temperature, suggests that the term R_{imp} in Eq. (2) is associated with a channel whose carrier concentration, n_{imp} has an exponential dependence with temperature, and a well-defined activation energy, ϵ_{imp} :

$$R_{imp} = \frac{r_{imp}}{en_{imp}} \propto \exp\left(\frac{\epsilon_{imp}}{k_B T}\right), \quad (3)$$

where e is the electron charge, k_B the Boltzmann constant, T the absolute temperature, and r_{imp} the Hall factor, which, although not known and possibly significantly different from unity in this almost-degenerate channel, remains constant in the whole low-temperature range. Assuming that this also holds at higher temperatures, we propose to extract R_{imp} by fitting R_H in the low temperature range, and then calculate R_{con} by inverting Eq. (2), provided we know how to split the total conductivity into the two components σ_{con} and σ_{imp} .

This information can be extracted by plotting the logarithm of the measured conductivities as a function of the inverse of the temperature, as shown in figure 2 for the samples of the Al_{0.6}Ga_{0.4}N series. It then becomes evident that the experimental data of the three samples are well fitted by a sum of two exponentials,

$$\sigma = \sigma_1 \exp\left(-\frac{\epsilon_1}{k_B T}\right) + \sigma_3 \exp\left(-\frac{\epsilon_3}{k_B T}\right), \quad (4)$$

where the activation energies ε_1 and ε_3 , and the asymptotic conductivities σ_1 and σ_3 are the fitting parameters. The former of the two terms, with subscript 1, accounts only for the band conduction and the slight deviation of the data from pure exponential dependence close to the 400 K limit is, most likely, due to an increased phonon interaction with the free carriers. The latter term (which following the convention of Shklovskii and Efros [14] is indicated with subscript 3) is attributed to nearest-neighbour hopping (NNH) conduction. The term usually indicated with the subscript 2 and attributed to the hopping among neutral donors is, as often the case, not detectable in our material.

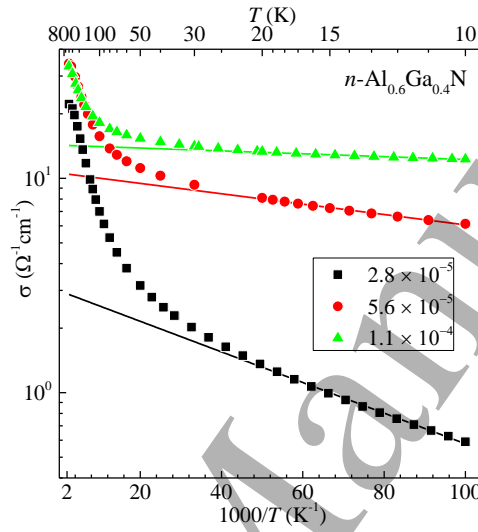


Figure 2. Temperature dependence of conductivity for the $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$ samples doped with different Si/III ratios. The straight lines show the exponential fitting at low temperatures extrapolated to higher temperatures.

Our attribution of the low-temperature conductivity to the NNH mechanism is based on the fact that the activation energy ε_3 remains extremely stable in this temperature range for each sample. This is a typical feature of this type of transport that can be explained as being due to the energy difference between the Fermi level and the peak of the density of states in the impurity band [14]. It is possible that in extending the analysis to lower temperatures, some form of variable-range hopping conduction might become apparent; but, for the scope of this study, this can be ignored without introducing any significant error in our model. Moreover, the observation that the low temperature conductivity increased by an order of magnitude (figure 2) when the dopant atoms supplied during growth increased by only a factor of two (from a Si/III ratio of 2.8×10^{-5} to 5.6×10^{-5} , before the channel became almost completely degenerate at 1.1×10^{-4}) is also consistent with hopping conduction [15]. In addition to that, the reduction of the activation energy with increased doping can be explained as the effect of an increased superposition among the localized state wavefunctions.

Potential fluctuations due to alloy disorder are also expected to be present in AlGaIn materials, and are known to induce tails of localized states into the gap. It is very possible that these states also contribute to the hopping transport observed in our material; however, because of the typical features of NNH, we believe that the low-mobility channel in our material is dominated by hopping in the impurity band.

As a result of those considerations, we can identify σ_{imp} in Eq. (2) with the second exponential term of Eq. (4), and because ϵ_3 is orders of magnitude lower than ϵ_1 , extract it by fitting the experimental data in the low temperature range (10–20 K); finally, by subtracting the so obtained σ_{imp} from the total conductivity we can obtain σ_{con} . It is important to notice that this procedure, which is justified by the NNH mechanism when a fixed activation energy ϵ_3 is present, also holds when ϵ_3 has completely vanished and the hopping conduction has become pure metallic transport in the degenerate impurity band, as it is almost the case for the sample with a Si/III ratio of 1.1×10^{-4} . It has been reported by Tasli *et al.* [16] that, in a completely degenerated $\text{Al}_{0.57}\text{Ga}_{0.43}\text{N}$ material, quantum corrections due to electron-electron interactions have to be included in the description of the low-temperature metal conductivity. Although it is likely that electron-electron interactions are important also in the insulator side of the transition here explored [14], this analysis would require measurements at lower temperatures and is outside the scope of this paper.

Apart from its use in retrieving R_{con} and R_{imp} from Eq. (2), the splitting of the conductivity into σ_{imp} and σ_{con} allowed us to extrapolate the contribution of the impurity-band channel at room-temperature, which was estimated to be 13%, 32% and 46% for the samples with 2.8×10^{-5} , 5.6×10^{-5} and 1.1×10^{-4} Si/III ratio, respectively. This result further confirms that the simple interpretation of our Hall-effect measurements based on a standard single-band approach is not reliable, even at room-temperature, and justifies the need for our method of separation.

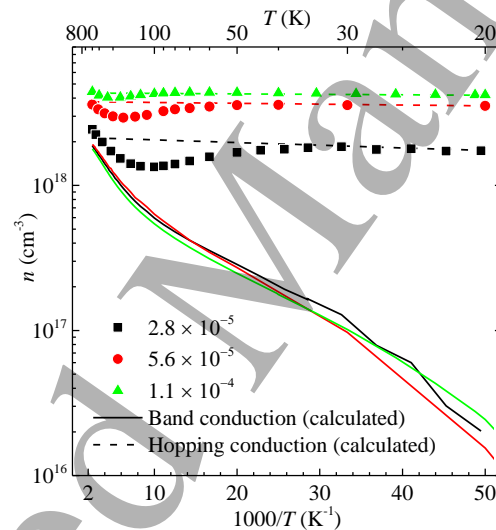


Figure 3. Temperature dependence of carrier concentration for the $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$ samples doped with different Si/III ratios. The discrete data-points represent the uncorrected carrier concentration values obtained with the standard single-channel model; the solid and dashed lines are the concentrations calculated with our model for the conduction-band and the impurity-band channel, respectively.

In figure 3 we present the separate values of the Hall carrier concentrations in the conduction-band and in the impurity-band channel as a function of temperature, which were obtained using the equations

$$n_{con, imp} = \frac{1}{eR_{con, imp}}. \quad (5)$$

For comparison, the combined effect of the two transport mechanisms interpreted with the standard single-channel model is also shown, with the apparent dip caused by the discrepancies in the mobilities of the two channels.

It is worth mentioning that these values have to be taken with care as we do not have any information about the Hall factors (r_{con} and r_{imp}), and while we can assume it is close to 1 for the diffusion transport in the conduction band, this might not be true for the impurity band of localized states. Nonetheless, even though this model is only meant to give an approximate estimation of R_{con} and R_{imp} , and it becomes inaccurate in the low temperature range where σ_{con} approaches zero and relative errors increase significantly, still a great deal of information can be extracted. In particular, our model suggests that although the uncorrected room-temperature carrier concentration appears to increase with the dopant concentration (up to $4.2 \times 10^{18} \text{ cm}^{-3}$ in the range explored) the actual free-carrier concentration in the conduction band has already reached its maximum of $1.6 \times 10^{18} \text{ cm}^{-3}$ at a Si/III ratio of 2.8×10^{-5} , and any further increase of dopant atoms supplied during growth only enhances the impurity-band conduction with concentrations of the carriers participating in this type of transport that increase from $2.1 \times 10^{18} \text{ cm}^{-3}$ up to $4.3 \times 10^{18} \text{ cm}^{-3}$. This process eventually stops when a critical Si/III ratio is reached and the band-like channel starts to degrade (i.e. at 2.2×10^{-4} and above; not shown here).

The free carrier concentrations in the conduction band, which, as shown in figure 3, are virtually the same in all the reported $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ samples, can be fitted using the charge neutrality equation:

$$\frac{n(n + N_A)}{N_D - N_A - n} = \frac{N_C}{g} \exp\left(-\frac{E_d}{kT}\right) \quad (6)$$

in which, apart from the symbols already defined, N_D and N_A are, respectively, the concentrations of donor and acceptor impurities, $g=2$ the ground state degeneracy, $N_C = (8.9 \times 10^{14}) \times T^{3/2}$ the effective density of states, and E_d the activation energy. This gives a very low E_d of about 1 meV, which is consistent with the presence of large screening effects due to the high concentration of free carriers, as already reported in literature [17-19]. Although the net dopant concentrations ($N_D - N_A$) in the three samples are most likely different, it was not possible to estimate them because, in this range of temperatures far from the saturation regime, the variations of n with $N_D - N_A$ are minimal. However the compensation degree (N_A/N_D) could be estimated as being around 27% for all the three samples.

Our model also allows the separation of the Hall mobilities of the two channels:

$$\mu_{con, imp} = \sigma_{con, imp} R_{con, imp} , \quad (7)$$

as shown, for the sample with a Si/III ratio of 2.8×10^{-5} , in figure 4. The trend is very similar in all the samples of the $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ series, with an impurity-band mobility almost constant in the range of temperature explored, and free-carrier mobility that shows the typical bell-shaped curve due to ionized impurity scattering dominating at lower temperatures and phonon scattering at higher temperatures.

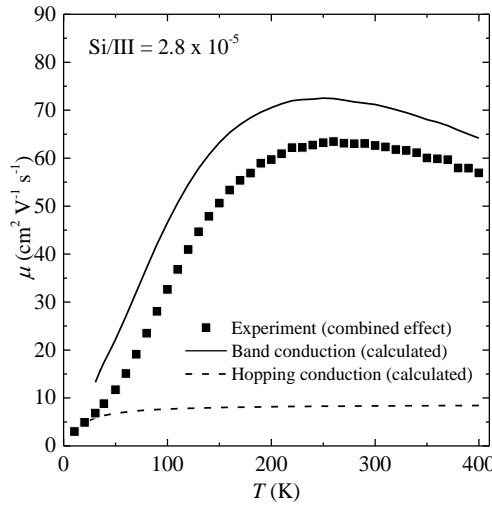


Figure 4. Typical Hall mobility for the samples of the $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$ series; the sample shown here has a Si/III ratio of 2.8×10^{-5} . The squares represent the uncorrected values obtained with the standard single-channel model; the solid and dashed lines are the Hall mobilities calculated with our model for the free carriers and the impurity-band channel, respectively.

4. Conclusion

We have demonstrated that in our MOVPE-grown $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$ material a substantial amount of the total conductivity is due to impurity-band conduction; in particular, the sample with lowest resistivity showed a room-temperature impurity-band transport fraction as high as 46% of the total. For the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{N}$ alloy composition we were not able to quantify its amount, but the presence of a maximum in the Hall coefficient proved that an impurity-band channel is present also in this material. We have proposed an approximate method to extract carrier concentrations and mobilities of both channels from Hall-effect measurements, which showed that an increase in doping $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ towards the optimum condition does not result in any change of the band-like conductivity component but still significantly increases the impurity-band part.

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